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# **FINAL TECHNICAL REPORT**

## **Grant AFOSR FA9550-04-1-0199**

### **Physics of Spin-Polarized Media**

by

William Happer and Yuan-Yu Jau

November 21, 2007

## **Overview**

The work supported by this AFOSR grant was focused on the fundamental physics and the applications of spin polarized species. Some of the most notable accomplishments were: (1) using optically pumped alkali-metal vapor to polarize the nuclei of solid materials; (2) making much simpler atomic clocks with the physics of push-pull optical pumping, an efficient new pumping method that we discovered during the course of the grant; (3) nonlinear “pressure shifts” of atomic clock frequencies due to the formation of van der Waals molecules; (4) the discovery and interpretation of unexpected signal reversals of magnetic resonance lines used in atomic clocks; (5) a new method of filling atomic clock and magnetometer cells by electrolysis through the glass walls; (6) new investigations of optical pumping and magnetic resonances of spin-polarized metastable xenon atoms; (7) the discovery of universal contaminants of the alkali metal used in atomic clocks and magnetometers.

Most of the work supported by this AFOSR grant has had and will continue to have applications to technologies of importance to the Department of Defense and to the United States Air Force. For example, atomic clocks based on push-pull pumping have substantially fewer parts and higher signal-to-noise ratios than conventional atomic clocks. They can be more stable, smaller, less expensive and consume less electrical power than conventional atomic clocks, and they could be used to improve the performance and decrease the cost of geolocation systems.

## **Major Accomplishments**

### **1. Spin-transfer optical pumping of solids**

Spin-transfer optical pumping of solids is a new technique, developed with the support of this grant. This is a non-cryogenic method that allows us to polarize nuclear spins in a solid by spin exchange with an optically pumped vapor that contacts the solid surface. A



similar method has been widely used to polarize the nuclei of various noble gases. In the first experiment of this type, we showed that optically pumped cesium vapor can be used to polarize  $^{133}\text{Cs}$  nuclei in CsH salt that coats the walls of a vapor cell. This result, an important first step, has been published in Physical Review Letters [1]. There we demonstrate that the polarization of nuclei  $^{133}\text{Cs}$  in CsH salt can be enhanced by at least a factor of 4 over the thermal-equilibrium value at a magnetic field of 9.4 tesla. Our experiments show that both the nuclear and electron spins of the optically pumped Cs atoms contribute to the spin transfer, as shown in Fig. 1. In subsequent studies, we have demonstrated polarization enhancement of Cs nuclei in CsCl and also the possibility of polarization enhancement of protons in CsH. The success of polarizing electrons or nuclei of different solids will lead to research applications in biology, materials physics, spintronics, etc. For example, nuclei of Li ions dissolved in liquid can have longitudinal spin relaxation times,  $T_1$ , of a few tens of seconds. Hyperpolarized lithium ions can therefore be used as a convenient tracer in biomedical research, microfluid systems, etc. Many proposed quantum computing systems would also work better with artificially enhanced spin polarization of the nuclei of solids. The detailed physics of how the spin transfer takes place is not yet understood in any detail, so we plan to continue work in this area.

## 2. Photonic clocks

Current approaches to miniaturize atomic clocks still require quartz-crystal local oscillators and associated frequency multiplication electronics and photodetectors, both of which contribute substantially to the cost and power requirements of the clock. During the course of this grant, we have successfully demonstrated a “laser-atomic oscillator.” The result has recently been published in Physical Review Letters [2]. This novel arrangement needs neither local oscillator with its associated electronics nor a photodetector to produce the clock signal. A cell with alkali-metal vapor, located inside the external optical cavity of a laser, and with passive optical elements that facilitate push-pull optical pumping, can cause the laser to spontaneously oscillate at the atomic clock frequency. The light is modulated at the 0–0 clock frequency, and it generates a comb of optical sidebands in the frequency domain, as illustrated in Fig. 2. To further improve their suitability for atomic clocks, atomic magnetometers, the generation of optical combs and other applications, we are continuing our studies of the basic physics and technical details of these systems.

## 3. Hyperfine frequency shifts due to van der Waals molecules

The most widely used atomic clocks are based on optically pumped Rb and Cs. In these clocks, an alkali-metal vapor is optically pumped by a lamp or a diode laser, and at the same time a hyperfine transition between ground-state sublevels is excited by magnetic resonance or by modulated light. In either case, it is necessary to use a chemically inert buffer gas to keep the pumped atoms from diffusing too quickly to the walls of the resonance cell. As a result of collisions of the gas atoms or molecules with the optically-pumped alkali-metal atoms, the clock frequency is shifted from the ideal hyperfine frequency, the Bohr frequency for the “0–0” transition between the two ground-state sublevels with azimuthal quantum numbers  $m = 0$ . The resulting “pressure shifts,” together with the “light shifts” of the clock



frequency are one of the limits on clock accuracy. To get the best possible clock performance, it is necessary to thoroughly understand the pressure shifts.

Practical clocks almost always contain a mixture of Ar and N<sub>2</sub> gas, with a mixing fraction that nearly cancels the temperature coefficient of the pressure shift. It has always been assumed that both gases cause a linear pressure shift, that is, if the partial pressure of the gas is doubled at the same temperature, the corresponding shift is doubled. Alkali-metal atoms are known to form loosely bound van der Waals molecules with Ar atoms. The molecules will also contribute to the shifts of the clock frequency, but the resulting shifts are expected to have substantial non-linear contributions at the low pressures that are used in practice. These non-linear shifts have been ignored in previous clock design. We have begun experiments to look for these non-linear shifts, to understand their physics in detail, and to assess the consequences of the nonlinear shifts on clock performance. Some preliminary experimental data from our work is shown in Fig. 3. This is the first experimental evidence that there really is a non-linear pressure shift, presumably coming from CsAr Van der Waals molecules. The nonlinear shift is very large compared to the level of precision needed for atomic clocks. We believe we can use our understanding of the basic physics of this newly discovered phenomenon to improve the performance of atomic clocks.

#### **4. Signal reversal of magnetic resonances**

From the studies of physics related to end-resonance atomic clocks [3] that were invented by our group, we have discovered an unusual effect, “a reversal of magnetic resonance lines.” In conventional magnetic-resonance spectroscopy of optically-pumped alkali-metal vapors, when the frequency of the microwave field is equal to one of the hyperfine Bohr frequencies of the atom, the vapor becomes more opaque, and there is a decrease in the intensity of the pumping light that passes through the vapor. We have found that under certain circumstances, just the opposite occurs, the vapor becomes more transparent at magnetic resonance, and more light passes through the cell to the photodetector. This curious signal reversal comes from interplay of optical pumping with nearly monochromatic, laser light with the effects of spin exchange collisions between alkali-metal atoms. We have developed a multilevel model that accounts for all of the details of our observations. The physics cannot be understood with the simplified three-level model that is so widely used in to analyze optical pumping of alkali-metal atoms. The results of this work have been recently published in Physical Review A [4].

#### **5. Electrolytic cell filling technique**

Getting free, unoxidized alkali metal into miniature anodically bonded cells has been a challenging problem for the development of miniature atomic clocks and atomic magnetometers. We have recently demonstrated a new method to introduce free alkali metal into anodically bonded, silicon-chip cells by electrolysis of alkali-metal ions through the glass window of the cell, with the silicon midsection of the cell serving as the cathode, and liquid molten salt outside the cell serving as the anode [5]. This new method is very promising for of mass production of cells fabricated on silicon wafers. During this grant period, we have demonstrated filling multiple cells on single silicon wafer with the electrolysis method.



## 6. Magnetic resonance for spin-polarized metastable xenon atoms

Metastable noble-gas atoms are closely analogous to alkali-metal atoms in their ground state. Both atoms have a loosely-bound valence electron in an  $ns_{1/2}$  orbital. For alkali-metal atoms, the non-valence electrons are in spherically symmetric, closed shells. For metastable noble-gas atoms, the non-valence not quite spherically symmetric, since there is a hole in the outermost  $p_{3/2}$  shell, which is missing the  $s_{1/2}$  valence electron. The spectrum of the atom is complicated by the interactions between the anisotropic core and the valence electron, but there remain many analogies to the spectrum of alkali-metal atoms.

In comparison to alkali-metal atoms, there are far fewer studies of metastable noble-gas atoms. Nevertheless, recent studies of cold atoms, atomic clocks, and atomic magnetometers, and hyperpolarized noble gas nuclei have stimulated the new interest in spin-polarized metastable noble-gas atoms. We have conducted a series of experiments to understand the physics of metastable xenon atoms in a sealed cell. We are currently investigating practical issues, like the lifetime of the discharged cell, as well as more fundamental issues like the broadening mechanisms of magnetic resonance linewidths. We have optically pumped  $^{129}\text{Xe}$  to produce highly spin-polarized metastable atoms, and we have observed magnetic resonances between hyperfine sublevels with resonance linewidths as small as 30 kHz. In light of these findings, we are increasingly optimistic that metastable noble gas atoms could be useful for atomic clocks and magnetometers. A big advantage of systems based on metastable noble gas atoms, compared to alkali-metal atoms, is that the metastable systems are very insensitive to the ambient temperature. This temperature insensitivity is one of the biggest advantages of optically pumped metastable  $^4\text{He}$  magnetometers over optically pumped alkali-metal magnetometers.

## 7. Contaminants of alkali metals.

During our studies of spin-transfer optical pumping, we discovered that the alkali-metal used to fill the cells was frequently contaminated by impurities that lowered the magnetic resonance frequency of the nuclei. We find that there are two, mutually exclusive types of impurities. One impurity has been unambiguously identified as oxygen, but the other is still unknown. These new discoveries may provide useful clues for how to prepare better vapor cells, and it may help us understand some of the cell and lamp aging problems that have been so troublesome for vapor cell atomic clocks and that have limited the lifetimes of GPS satellites. The results of this study were published in Physical Review Letters [6].

## References

- [1] K. Ishikawa, B. Patton, Y.-Y. Jau and W. Happer, Spin transfer from an optically pumped alkali vapor to a solid, Phys. Rev. Lett. **98**, 183004 (2007).
- [2] Y.-Y. Jau and W. Happer, Push-pull Laser-Atomic Oscillator, Phys. Rev. Lett. in press (2007).
- [3] Y.-Y. Jau, A. B. Post *et al.*, Intense, Narrow Atomic-Clock Resonances, Phys. Rev. Lett. **92**, 110801 (2004).

- [4] F. Gong, Y.-Y. Jau, W. Happer, Magnetic resonance reversals in optically pumped alkali-metal vapor, *Phys. Rev. A* **75**, 053415 (2007).
- [5] F. Gong, Y.-Y. Jau, K. Jensen and W. Happer, Electrolytic fabrication of atomic clock, *Rev. Sci. Instrum.* **77**, 076101 (2006).
- [6] B. Patton, K. Ishikawa, Y.-Y. Jau and W. Happer, Intrinsic impurities in glass alkali-vapor cells, *Phys. Rev. Lett.* **99**, 027601 (2007).

## Personnel

AFOSR's support for the training of students and postdoctoral research associates is one of the most important benefits of the grant to Princeton University and to the United States. The following personnel have received full or partial support from Grant AFOSR FA9550-04-1-0199:

Dr. William Happer, Principal Investigator.

Dr. Yuan-Yu Jau, Instructor.

Dr. Brian Patton, Research Associate.

Dr. Steven Morgan, Research Associate.

Dr. Nick Kuzma, Research Associate.

Dr. Eli Miron, Visiting Scholar.

Dr. Kiyoshi Ishikawa, Visiting Scholar.

Ms. Fei Gong, Graduate Student.

Mr. Xia Tian, Graduate Student.

Mr. Ben Olsen, Graduate Student.

Ms. Amber Post, Graduate Student (received PhD degree in 2005, now at UVA).

Ms. Kate Jensen, Undergraduate Student (Princeton Class of 2005, now at MIT).

Mr. Paul Oreto, Undergraduate Student (Princeton Class of 2004, now at Harvard).

## Publications

Listed below are publications resulting from work with full or partial support from AFOSR.

1. P. J. Oreto, Y. -Y. Jau, A. B. Post, N. N. Kuzma and W. Happer, Buffer-Gas Induced Shift and Broadening of Hyperfine Resonances in Alkali-Metal Vapors, *Phys. Rev. A* **69**, 042716 (2004).

2. Y.-Y. Jau, A. B. Post, N. N. Kuzma, A. M. Braun, M. V. Romalis and W. Happer, Intense, Narrow Atomic Clock Resonances, *Phys. Rev. Letters* **92**, 110801 (2004).
3. Y.-Y. Jau, E. Miron, A. B. Post, N. N. Kuzma and W. Happer, Push-Pull Optical Pumping of Pure Superposition States, *Phys. Rev. Letters* **93**, 160802 (2004).
4. A. B. Post, Y.-Y. Jau, N. N. Kuzma and W. Happer, Amplitude- Versus Frequency-Modulated Light for Coherent Population Trapping Resonances at High Buffer-Gas Pressures, *Phys. Rev. A* **72**, 033417 (2005).
5. Y.-Y. Jau and W. Happer, Simultaneous Stabilization of Frequency and Magnetic Field with the Tilted 0-0 State, *Applied Physics Letters* **87**, 204108 (2005).
6. F. Gong, Y.-Y. Jau, K. Jensen and W. Happer, Electrolytic Fabrication of Atomic Clock Cells, *Rev. Sci. Instrum.* **77**, 076101 (2006).
7. K. Ishikawa, B. Patton, Y.-Y. Jau and W. Happer, Spin Transfer from an Optically Pumped Alkali Vapor to a Solid, *Phys. Rev. Letters* **98**, 183004 (2007).
8. F. Gong, Y.-Y. Jau and W. Happer, Magnetic Resonance Reversals in Optically Pumped Alkali-Metal Vapor, *Phys. Rev. A* **75**, 053415 (2007).
9. B. Patton, K. Ishikawa, Y.-Y. Jau and W. Happer, Intrinsic Impurities in Glass Alkali-Vapor Cells, *Phys. Rev. Letters* **99**, 027601 (2007).
10. Y.-Y. Jau and W. Happer, Push-pull Laser-Atomic Oscillator, *Phys. Rev. Letters*, in press (2007).



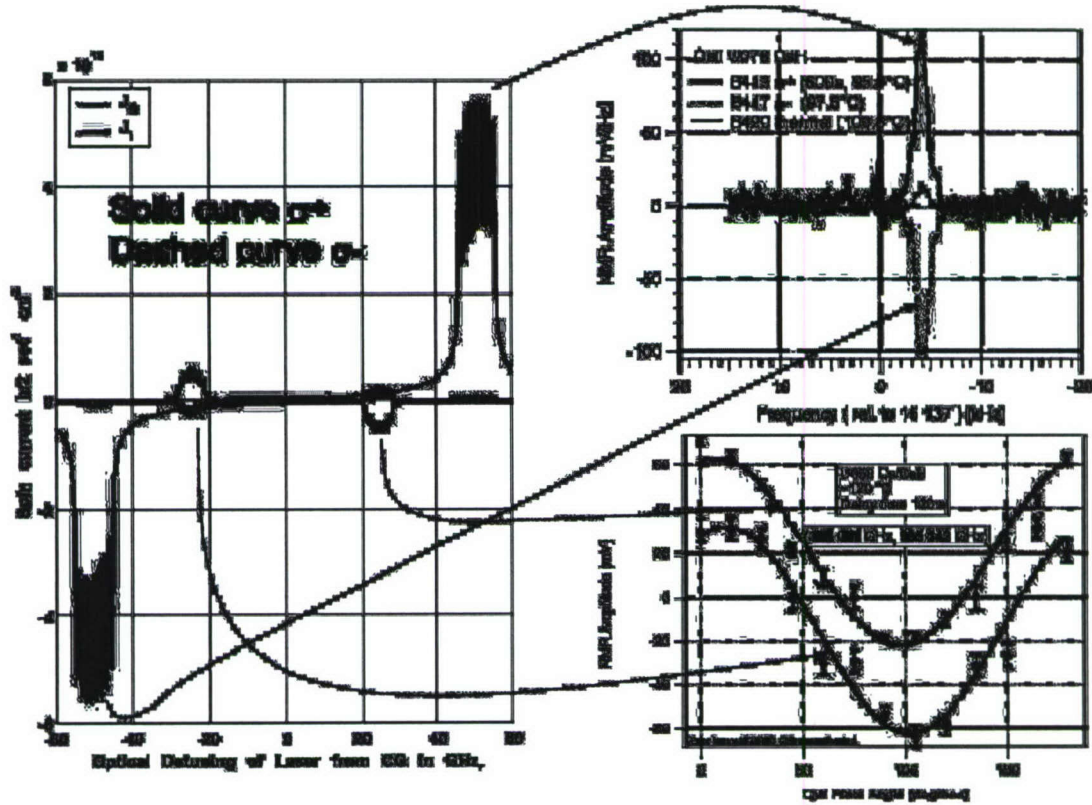


Figure 1: The top-right panel shows that the NMR signals (red traces) of Cs nuclei in CsH salt that is exposed to optically pumped Cs vapor are enhanced by a factor of 12 compared to the thermal NMR signal (black trace) when the vapor is not optically pumped. The left panel shows that by appropriate tuning of the laser wavelength, we can ensure that the major carrier of angular momentum from the optically pumped vapor to the surface is either the electron spin current  $J_S$  or the nuclear spin current  $J_I$ , modelled as red and blue lines. The bottom-right panel shows that the nuclear spin polarization of Cs vapor makes a significant contribution to the CsH NMR signals.



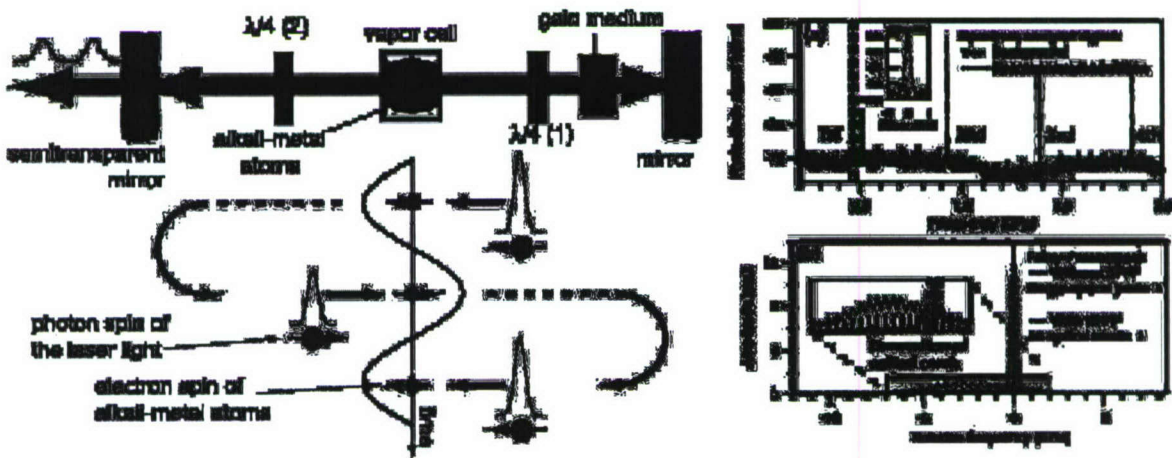


Figure 2: Left panel: key components of a push-pull laser-atomic oscillator. The circularly polarized laser pulses spontaneously synchronize with the electron spin oscillation of the atomic vapor inside the laser cavity. Right panel: experimental data showing the power spectrum of pulsed light and the comb structure (sidebands) of the the laser's optical spectrum. Neither a quartz crystal local oscillator, nor a photodetector are needed for this atomic-clock configuration. A strong clock signal can be retrieved from the spontaneous microwave modulation of the voltage across the laser diode.

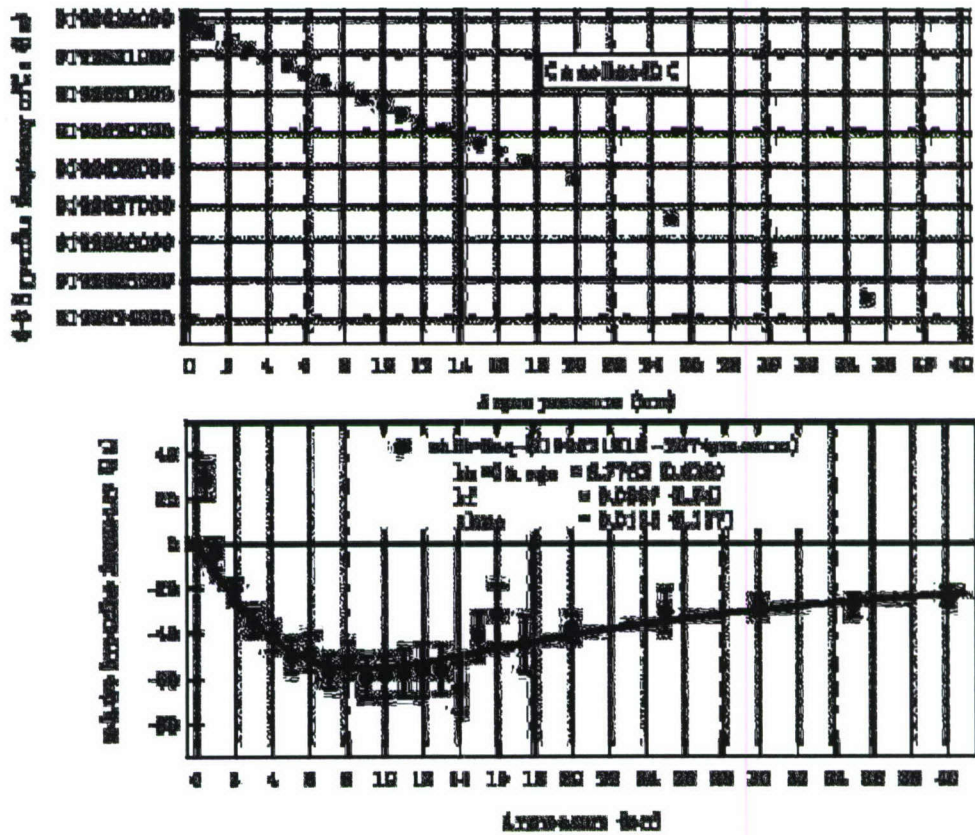


Figure 3: Top panel: the Cs hyperfine frequency as a function of the argon buffer-gas pressure. Bottom panel: after subtraction of the linear, limiting shift at high pressures, a substantial nonlinear contribution to the frequency shift at lower pressures is revealed.